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STUDY OF NONEQUILIBRIUM FLOWS

J. W. Rich

Cornell Aeronautical Laboratory, Inc.  
Buffalo, New York

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CORNELL AERONAUTICAL LABORATORY, INC.  
Buffalo, New York 14221

Final Summary Report

STUDY OF NONEQUILIBRIUM FLOWS

by: J. W. Rich

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# STUDY OF NONEQUILIBRIUM FLOWS

## FINAL SUMMARY REPORT

Contract No. AF 49(638)-1488

December 1, 1964 - December 1, 1967

This report presents a brief summary of work performed under contract AF 49(638)-1488 over the three-year period December 1, 1964 to December 1, 1967. Included are the abstracts of reports and papers published under this contract. The most recent experimental data, which have not as yet been published, are also given here.

### 1. RESEARCH OBJECTIVES

Research under this contract has been concerned with theoretical and experimental studies of thermal nonequilibrium which can occur in high stagnation-enthalpy flows of current technical interest. Specifically, the emphasis has been on obtaining a detailed description of the kinetic mechanisms involved in the vibrational relaxation of diatomic molecules of current interest in both vibrational excitation (shock-wave) and vibrational deexcitation (expansion-flow) environments. To this end, the experimental part of the project has concentrated on development and utilization of various spectroscopic diagnostic techniques for monitoring vibrational state populations in high-temperature relaxing gas flows. The theoretical part of the project has centered on master-equation analyses of various models for systems of vibrationally relaxing molecules.

## 2. BACKGROUND

Under an earlier AFOSR contract (AF 49(638)-792 ), measurements of  $N_2$  vibrational relaxation were made in high-enthalpy nozzle expansion flows<sup>1</sup> and in flows behind normal shock waves.<sup>2</sup> For this purpose, the technique of spectrum line reversal<sup>3</sup> was adopted for use in such flows. A major finding of these investigations was that the expansion flow measurements indicated a much shorter relaxation time than that measured at a comparable temperature behind a shock wave. It also was found that the line-reversal measurements of relaxation time behind a shock wave agreed with times measured behind shocks using more conventional techniques. The experimental results apparently indicated a basic difference between the vibrational excitation and vibrational deexcitation processes. Further work<sup>4</sup> under a related contract indicated that the anomalously fast relaxation times in  $N_2$  expansion flows were not the result of impurity effects.

Subsequent to the expansion flow work done at CAL, the existence of fast expansion flow relaxation times in  $N_2$  and in air was confirmed by measurements made in other laboratories.<sup>5, 6, 7, 8</sup> An encouraging aspect of these other experiments was that they were performed in differing flow configurations and, in some cases, using a different measurement technique (electron beam) than that used in the CAL experiments.

The growing body of experimental evidence pointing toward a difference in the nature of the vibrational excitation and deexcitation process made theoretical investigation desirable. The observed faster expansion flow relaxation times could not be interpreted using existing theory. Furthermore,

entirely apart from unexplained experimental results, existing theory did not adequately treat certain features present in the relaxation of real diatomic molecules, such as molecular anharmonicity and vibration-vibration (V-V) exchange processes. One aspect of research under the present contract, therefore, has centered on analysis of more exact theoretical models of the relaxation process.

The experimental phase of the present contract has concentrated on development of a more direct means of measuring the vibrational state populations in relaxing flows. As mentioned above, both spectrum line-reversal techniques and the electron beam method<sup>5,6,8</sup> have yielded mutually confirmatory measurements of vibrational relaxation rates in expanding flows. The agreement obtained by using two such widely different means of measurement was quite encouraging; it did not appear likely that the two methods would show similar measured rates, if significant errors were present. Both methods are, however, quite indirect. They require accurate quantitative knowledge of fairly complex molecular processes other than the one under study. For this reason, a spectrophotometric method of following the populations of the  $V=1$  state of infrared active diatomics was developed under the present contract.

### 3. WORK UNDER THIS CONTRACT

#### a.) Theoretical

Beginning in the latter part of 1965, a theoretical study was initiated to examine the basic analytical framework for describing the vibrational relaxation process. The motivation for this work was a desire to reconcile the shock-

wave and expansion-flow vibrational relaxation rates for  $N_2$  within a single theory.

The first analysis performed under this contract investigated the results of including molecular anharmonicity effects in the vibration-translation (V-T) transition probabilities. The first phase of the work was confined to the case of diatomics relaxing in a heat bath of inert diluent; this work was reported at the 1965 meeting of the Fluid Dynamics Division of the American Physical Society (abstract 1).<sup>9</sup> The work was extended to a pure gas model by including effects of vibration-vibration (V-V) exchange transition probabilities; a detailed presentation was published in the proceedings of the 11th International Symposium on Combustion (abstract 2).<sup>10</sup> It was concluded, on the basis of this work, that inclusion of anharmonicity effects only in the V-T model probabilities could not, by itself, explain the experimental observations.

In 1967, work by C. E. Treanor<sup>11</sup> at CAL on the effect of non-resonant V-V exchange among anharmonic oscillators was incorporated into the formalism previously developed<sup>10</sup> under this contract. Several additional facets of the relaxation model were also analyzed. The results of this work were made available as an AFOSR technical report (abstract 3)<sup>12</sup> and also as part of a paper which will be published in the Journal of Chemical Physics (abstract 4).<sup>13</sup> A major result of this investigation was that the theory developed provided a plausible interpretation of the contrasting behavior of vibrational excitation (shock wave) processes and vibrational deexcitation (expansion) processes. Detailed experimental verification of many predictions of the theory remains, however, a task for the future.

#### b.) Experimental

During 1965 modifications to the basic experimental apparatus were effected in order to extend the scope of experimental investigation. First, the spectroscopic line-reversal apparatus was redesigned in order to improve spatial resolution and facilitate simultaneous multichannel reversal measurements of vibrational temperatures. With the improved system, good signal-to-noise ratios were obtained with small beam diameters. Secondly, the shock tube was extended from 20 ft to 36 ft in order to increase the available test flow duration for present operating conditions and to permit operation over a wider range of nozzle reservoir temperatures with sufficient nozzle-flow test duration to obtain accurate diagnostic measurements. In addition, provision also was made for obtaining simultaneous line-reversal temperature measurements behind the incident shock wave as well as in the nozzle flow. With these additions it was possible to observe vibrational relaxation processes in excitation and deexcitation environments in a single experiment.

Experiments were then conducted to investigate the possibility of utilizing the Na resonant doublet at  $3300 \text{ \AA}$  for line reversal measurements. Such measurements are useful for observing the relaxation of higher vibrational levels, about the 13th vibrational level for  $N_2$ . Preliminary experiments were conducted behind the incident shock wave by monitoring the Na emission at  $3300 \text{ \AA}$  using a small prism spectrograph and the Na emission at  $5893 \text{ \AA}$  using an interference filter. A comparison of these results indicated adequate signal-to-noise ratio to permit reversal measurements at  $3300 \text{ \AA}$ .

The limited experimental effort during 1966 was concerned chiefly with the reexamination of previous shock wave data for  $N_2$  vibrational relaxation, an attempt to measure  $N_2$  vibrational relaxation rates much closer to the shock front (where departures from equilibrium are largest) than had heretofore been attempted, and with the study and design of a more direct spectroscopic technique involving the infrared spectral region for vibrational temperature measurement.

With respect to the studies closer to the shock front, Na line-reversal measurements of the  $N_2$  vibrational temperatures were obtained in the region from about 1 microsecond to about 10 microseconds after passage of the shock front past the observation station. In several of these measurements, reversal temperatures were obtained over a period in which there was a significant decrease in translational temperature during the relaxation process. From such data it was possible to determine not only the vibrational relaxation rate but the dependence of the rate on translational temperature as well.

The mean value of the relaxation rates over the relaxation period were, in general, found to agree very well with previous measurements. However, the dependencies of these rates on translational temperature differed from the usual Landau-Teller form,  $\exp(-T^{-1/3})$ . The significance of these results was somewhat comprised by the uncertainty in the rates for the dissociation of the NaCl additive. In view of this uncertainty, further investigation of the near shock region was deferred until after development of the infrared diagnostic method.

In accordance with the above considerations, a study was made during 1966 of the application of the reversal technique to the infrared spectral



region, in which CO is used as the thermometric element. (The coupling of CO and N<sub>2</sub> vibration has been shown to be very large so that rapid equilibration is assured.)

During the latter part of 1966 and 1967 the infrared band reversal technique was developed by measurement of vibrational temperature in CO under support of NASA (contract NASr-109). However, the background-radiation source essential for absorption measurements was developed under the present contract. The considerable effort expended in the development of this light source resulted in a configuration which represents a suitable compromise among the requirements of high brightness temperature, long life, and easy replacement of the radiating element. The present configuration of the radiation source consists of a 1/4-in. O.D. x 3-in. long spectroscopic-carbon rod which is bored internally to about 1/8 in. The central inch of the element is further machined externally to give a wall thickness of 0.015-0.020 in. This element is heated resistively in an argon atmosphere; the electrical connections are made through solid carbon pieces which are tightly fitted to each end of the 3-in. cylindrical element. Further, one wall of the radiation element has a 0.060-in. hole drilled radially at its midpoint to expose the inner core of the element. The entire assembly is then supported in a water-cooled housing. With this configuration, temperatures of about 2400-2600°K (as measured with an optical pyrometer at 6550 Å) have been obtained for a current of about 120 amperes at 12 volts.

Under the support of the NASA contract, the infrared diagnostic technique, using this background source, was applied to the study of the vibrational relaxation of CO itself in shock-wave and nozzle-expansion flows.

The results of these studies<sup>14</sup> showed that the technique does provide a reliable and sensitive method for measuring the vibrational temperatures of CO. In particular, the vibrational relaxation rates obtained in shock-wave flows were found to agree well with those obtained by other investigators using other diagnostic techniques. Moreover, the expansion-flow results completely supported the occurrence of faster vibrational de-excitation rates in nozzle flows than those predicted on the basis of the shock-wave results, as was initially observed for N<sub>2</sub> under this contract.

With the favorable completion of these CO studies, experiments with N<sub>2</sub> were initiated. At present, experiments have been completed for shock-wave flows. A number of measurements of the relaxation rate of N<sub>2</sub> have been obtained behind shock-wave flows in the temperature (equilibrium) range of about 1500 to 2100°K and pressures near 1 atmosphere. These rates determined by the CO infrared band-reversal method are shown in Fig. 1, where the product of the relaxation time ( $\tau_s$ ) and the pressure (P) is plotted against the equilibrium temperature behind the shock wave. This product provides a pressure-normalized relaxation time, and the results are in very good agreement with other published data for the vibrational relaxation of N<sub>2</sub> behind shock waves. This agreement among the rates attests to the applicability of CO as a thermometric additive for measurements of N<sub>2</sub> vibrational temperatures.

Some preliminary measurements of the vibrational temperature of N<sub>2</sub> in nozzle flows using this infrared technique have also been obtained. During the course of these experiments, the graphite-element background radiation source required for the absorption measurements burned out

and a replacement was fabricated. As is the case for such a replacement, the effective brightness of the radiation source in the infrared was rechecked by utilizing the equilibrium temperature region behind incident shock waves. Subsequently, the InSb infrared detector failed, necessitating a suspension in these experiments pending the receipt of a new detector.

The available nozzle-flow data are shown in Fig. 2. The open symbols represent the results obtained previously under this contract using Na line-reversal spectroscopy. The four sets of data points shown by the filled-in symbols are the present infrared data. The upper point for each reservoir temperature indicates the measured CO vibrational temperature, while the lower point represents the  $N_2$  vibrational temperature which is calculated from the former on theoretical grounds.<sup>11</sup> The infrared results are clearly in good agreement with the line-reversal data and hence confirm the faster relaxation of  $N_2$  vibration in nozzle flows than in shock-wave flows. The curves labeled  $\tau/\tau_s$  are calculated using a value of the vibrational relaxation time ( $\tau$ ) equal to  $10^{-1}$  or  $10^{-2}$  of the value  $\tau_s$  given by the fitted shock-wave curve of Ref. 2.

It is hoped to obtain a few more data for the nozzle flows upon receipt of the new detector, thereby concluding this phase of the research.

The experimental work done under this contract to May 1966, forms part of a paper presented at the 7th AGARD Propulsion and Energetics Panel Colloquium in Oslo (Abstract 5).<sup>(15)</sup>

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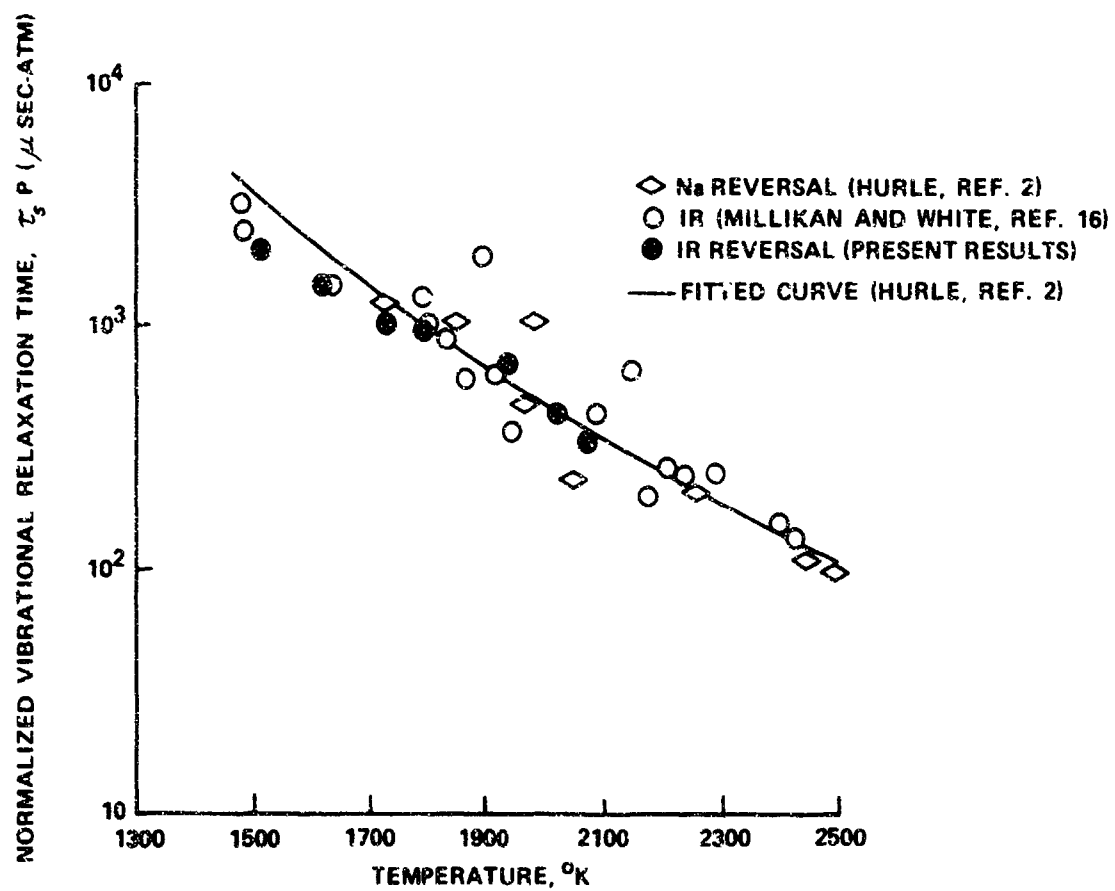


Figure 1 NORMALIZED VIBRATIONAL RELAXATION TIMES,  $\tau_3 P$ , FOR  $N_2$  FROM SHOCK-WAVE EXPERIMENTS

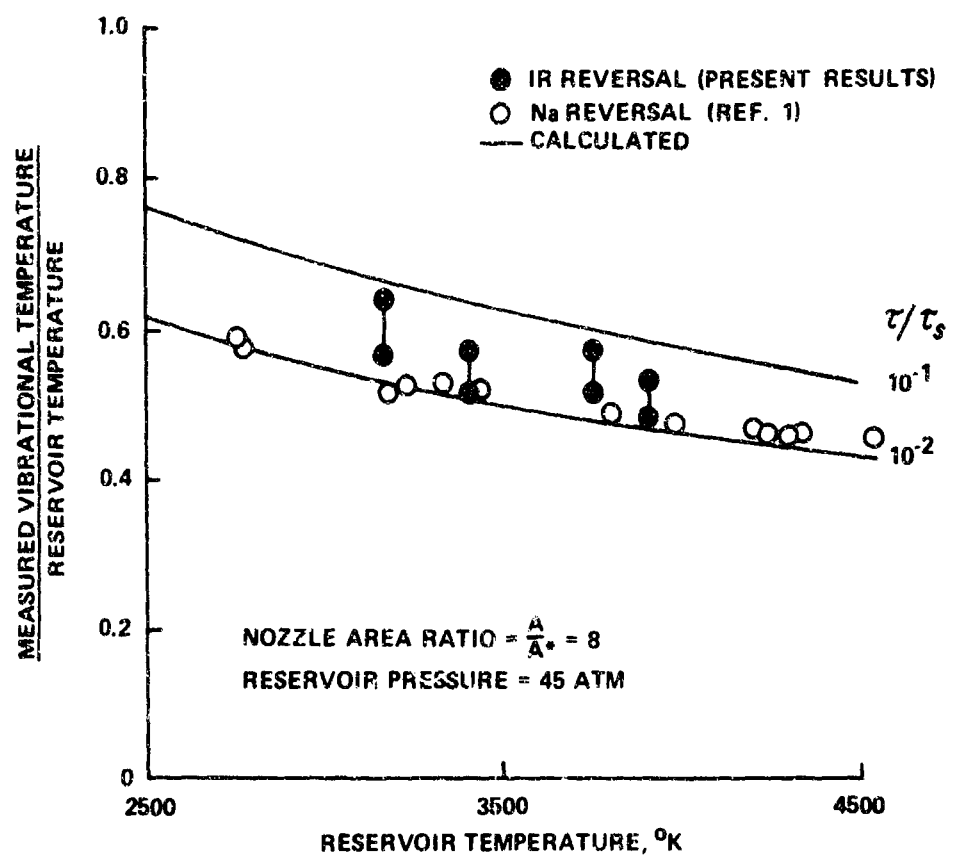


Figure 2 VIBRATIONAL TEMPERATURES FOR  $N_2$  MEASURED IN EXPANSION FLOWS

ABSTRACTS OF PAPER AND REPORT PUBLICATIONS  
BASED ON RESEARCH PERFORMED ON CONTRACT AF 49(638)-1488  
WITH THE AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

ABSTRACT NO. 1

Paper B-7, Division of Fluid Dynamics, American Physical Society,  
Cleveland, Ohio, 22-24 November 1965.

A MASTER EQUATION SOLUTION FOR VIBRATIONAL RELAXATION\*

J. W. Rich (introduced by A. Hertzberg)  
Cornell Aeronautical Laboratory, Inc.

The relaxation of a system of oscillators in contact with a constant temperature heat bath is considered. The model is the same as that adopted by Rubin and Shuler,<sup>1</sup> save that collisional transition probabilities are allowed to have arbitrary exponential dependence on initial quantum number, i. e.,  $P_{n, n+1} = P \exp [(n+1) a]$ . Solution is obtained in the high temperature limit,  $\theta/T \ll 1$ . The population of vibrational states as a function of time is expressed in terms of Bessel functions. It is found that the upper states equilibrate to the heat bath temperature much more rapidly than the lower states. Previous experimental results from high-temperature expansion flows, in which the population of excited vibrational states is monitored, are discussed in terms of the present analysis. These results are consistent with the interpretation that upper states are relaxed much more rapidly than lower states.

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<sup>1</sup>Rubin, R. J. and Shuler, K. E., J. Chem. Phys. 25, 59, 68 (1956).



## ABSTRACT NO. 2

Presented at Eleventh Symposium (International) on Combustion, Berkeley, California, 14-20 August 1966 (also CAL Report No. AF 2022-A-1, April 1966)

### POPULATION DISTRIBUTIONS DURING VIBRATIONAL RELAXATION OF DIATOMIC GASES

J. W. Rich and R. G. Rehm  
Cornell Aeronautical Laboratory, Inc.

A master equation analysis for the vibrational relaxation of a diatomic gas is presented, including the effects of near-resonance exchange of vibrational energy between colliding oscillators. It is found that the mechanism of rapid near-resonance exchange of vibrational energy among the diatomic species tends to maintain a quasi-steady state vibrational population distribution during the relaxation of a pure gas. The specific functional form of this quasi-steady-state distribution is determined from a perturbation analysis of the master equation. The remainder of the paper gives detailed analyses of the relaxation equations for the case of equi-distant vibrational energy-level spacing. The analyses use a simple exponential form for the vibration-translation transition probabilities that preserves some degree of arbitrariness, and allows the probabilities to increase with quantum number at a rate which can differ significantly from the Landau-Teller expressions. It is found that when the diatomic gas is diluted by an inert to the extent that oscillator-oscillator exchange collisions no longer influence relaxation, the vibrational population distribution can depart markedly from the Boltzmann form. Whenever exchange collisions predominate, however, it is found that a near-Boltzmann distribution is preserved, regardless of the form of the vibration-translation transition probabilities, and the energy relaxation equation does not differ greatly from the Landau-Teller result.

### ABSTRACT NO. 3

AFOSR Scientific Report AFOSR 67-1180, also CAL Report No. AF 2022-A-2  
(May 1967)

#### VIBRATIONAL RELAXATION OF ANHARMONIC OSCILLATORS

J. W. Rich and R. G. Rehm  
Cornell Aeronautical Laboratory, Inc.

The terms in the master equation for vibrational relaxation of anharmonic oscillators are ordered according to the rates of the relaxation processes (vibrational exchange, vibrational-energy transfer to translation). A perturbation procedure is developed by making use of the fact that the vibration-translation energy exchange transitions are much slower than the vibration-vibration transitions. The population distributions in the master equation are expanded about their values when the vibration-vibration mechanism is the only one present. To zeroth order, the master equation is satisfied by a distribution derived previously by Treanor for anharmonic oscillators. This distribution is maintained by vibration-vibration exchange, and is only Boltzmann during relaxation in the limit of vanishing anharmonicity. The perturbation on this distribution, created by vibration energy transfer to translation, is calculated to first order for a special case.

The relaxation of the first moment of the zeroth-order distribution function is also investigated. It is found that, in the majority of cases, the effect of anharmonicity on vibrational relaxation is slight, and the system behaves in a fashion similar to the Landau-Teller model of vibrational relaxation. However, a significant exception occurs when the translational temperature is quite low, and the vibrational energy is very large. Under these conditions, the vibrational energy exchange mechanism creates a slight inversion in the vibrational population distribution. Moreover, for these conditions, the population of the lowest vibrational states can be considerably lower than the population predicted by the Landau-Teller model for the relaxation process. Furthermore, since the inversion tends to weight those states having fast vibration-to-translation energy transfer rates, the overall energy relaxation rate can be accelerated.

#### ABSTRACT NO. 4

Paper to be published in the Journal of Chemical Physics.

#### VIBRATIONAL RELAXATION OF ANHARMONIC OSCILLATORS WITH EXCHANGE-DOMINATED COLLISIONS

C. E. Treanor, J. W. Rich, and R. G. Rehm  
Cornell Aeronautical Laboratory, Inc.

The terms in the master equation for vibrational relaxation of anharmonic oscillators are ordered according to the rates of the relaxation processes (vibrational exchange, vibrational-energy transfer to translation). The population distributions in the master equation are expanded about their values when the vibration-vibration mechanism is the only one present. An analytic expression is given for the distribution maintained by the vibration-vibration mechanism. In the limiting case of the simple harmonic oscillator, this distribution reduces to the usual Boltzmann-like distribution defined by a single vibrational temperature. The general solution also applies to a mixture of simple-harmonic-oscillator gases of different fundamental frequencies. For such a mixture, each gas relaxes in a Boltzmann-like distribution, but the different gases have different (but related) vibrational temperatures at any given time.

The relaxation of the first moment of the distribution function also has been investigated. Anharmonicity causes a marked departure from the Landau-Teller model of vibrational relaxation under conditions of high vibrational energy, coupled with low translational temperature. For such conditions, the populations of the lower vibrational states can be considerably lower than those predicted by the Landau-Teller model. Furthermore, the overall energy relaxation rate can be accelerated.

## ABSTRACT NO. 5

Presented at the Seventh AGARD Colloquium on Recent Advances in Aero-thermochemistry, Propulsion and Energetics Panel, Oslo, Norway, 1966. (AGARD Conference Proceedings No. 12, p. 443, Ed. I. Glassman)

### RECENT STUDIES OF NONEQUILIBRIUM FLOWS AT THE CORNELL AERONAUTICAL LABORATORY J. Gordon Hall and Anthony L. Russo Cornell Aeronautical Laboratory, Inc.

The present paper reviews recent work on three problems concerned with nonequilibrium flows carried out in the Aerodynamic Research Department of the Cornell Aeronautical Laboratory. The first problem, discussed in Section 2, is that of vibrational relaxation in supersonic nozzle flows of diatomic gases. The initial line-reversal temperature measurements of Hurle, Russo, and Hall<sup>1</sup> for nozzle flows of  $N_2$ , which showed unexpectedly fast vibrational relaxation, are first briefly reviewed. Recent confirmatory studies, at other laboratories as well as at CAL, are then described. The results of different investigators for air as well as  $N_2$  are correlated. Line-reversal temperature data recently obtained for nozzle expansions of CO are presented which show fast vibrational relaxation similar to  $N_2$  expansions.

The second problem, discussed in Section 3, concerns nonequilibrium effects occurring in high-enthalpy air flows over thick wedge-flat plate bodies where strong (Prandtl-Meyer) flow expansion occurs at the wedge-plate corner. Shock-tunnel experiments and complementary nonequilibrium flow analyses carried out by Vidal and Stoddard<sup>2</sup> on this problem are briefly described. The experiments indicated substantial nonequilibrium effects on the flat-plate surface pressures. Boundary-layer displacement effects were found to be large following the corner expansion and limited the quantitative interpretation of the data.

The third problem, discussed in Section 4, is that of calculating the inviscid nonequilibrium flow field about an Apollo-type re-entry body at

ABSTRACT NO. 5 (cont.)

arbitrary angle of attack. A time-dependent direct method for this problem is briefly described which has been recently developed by Bohachevsky and Mates.<sup>3, 4</sup> Application by Dunn et al<sup>5</sup> of the ideal-gas solution from this method to estimate nonequilibrium ionization in the Apollo plasma sheath is discussed. Typical results are described for electron density distribution and consequent electromagnetic signal attenuation.

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13 ABSTRACT <p>This report presents a brief summary of work performed under contract AF 49(638)1488 over the three-year period December 1, 1964 to December 1, 1967. The experimental part of the project has concentrated on development and utilization of various spectroscopic techniques for monitoring vibrational state populations in high-temperature relaxing gas flows. The theoretical part of the project has centered on master-equation analyses of various models for systems of vibrationally relaxing molecules. Abstracts of reports and papers published under this contract are included. The most recent experimental data, which have not as yet been published, are also given here.</p>		

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